

What is claimed is:

1. A method for producing a membrane-electrode structure, comprising the steps of:

applying on a sheet substrate a catalyst paste comprising an electron conducting material supporting a catalyst and an ion conducting material, and drying the same, so as to form a electrode catalyst layer;

thermally transferring said electrode catalyst layer onto each side of a polymer electrolyte membrane, so as to form a laminated body such that said electrode catalyst layer is connected to each side of said polymer electrolyte membrane;

applying on a carbon substrate layer a first slurry comprising a water-repellent material and an electron conducting material and drying the same to form a water-repellent layer, and then applying on said water-repellent layer a second slurry comprising an electron conducting material and an ion conducting material and drying the same to form a hydrophilic layer, so as to form a diffusion electrode consisting of said carbon substrate, said water-repellent layer and said hydrophilic layer; and

laminating the previously formed diffusion electrode on said electrode catalyst layer of said laminated body through said hydrophilic layer, pressing the two under heating, so as to integrate said laminated body and said diffusion electrode.

2. The method for producing a membrane-electrode structure according to claim 1, wherein said second slurry comprises a pore-forming material.

3. The method for producing a membrane-electrode structure according to claim 1, wherein said catalyst paste comprises a pore-forming material.

4. The method for producing a membrane-electrode structure according to claim 1, wherein each of said catalyst paste and said second slurry comprises a pore-forming material, and wherein said hydrophilic layer and said electrode catalyst layer are formed such that the ratio of the volume of pores with a pore size of 0.01 to 1  $\mu\text{m}$  formed in said electrode catalyst layer by said pore-forming material to the volume of pores with a pore size of 0.01 to 1  $\mu\text{m}$  formed in said hydrophilic layer by said pore-forming material is less than 1.0.

5. The method for producing a membrane-electrode structure according to claim 1, wherein said hydrophilic layer and said electrode catalyst layer are formed such that the ratio of the weight of an ion conducting material contained in said electrode catalyst layer to the weight of an ion conducting material contained in said hydrophilic layer is set within the range of 1.0 to 1.4.

6. The method for producing a membrane-electrode structure according to claim 1, wherein said hydrophilic layer and said

electrode catalyst layer are formed such that the ratio of the weight of solid content in said electrode catalyst layer to the weight of solid content in said hydrophilic layer is set within the range of 1.0 to 3.5.

7. The method for producing a membrane-electrode structure according to claim 1, which comprises applying said second slurry on said water-repellent layer and drying the same, so as to form a hydrophilic layer having the maximum height of surface roughness,  $R_{max}$  of 40  $\mu\text{m}$  or less.

8. The method for producing a membrane-electrode structure according to claim 7, wherein said hydrophilic layer is formed such that it has a surface roughness in which the ratio of the surface area to the unit area is 1.25 or less.

9. The method for producing a membrane-electrode structure according to claim 7, wherein said water-repellent layer and said hydrophilic layer are formed such that the differential pressure between one side of said diffusion electrode and the other side thereof is set within the range between 100 and 300 mmAq, when the air is supplied at a flow rate of  $0.5 \text{ L/cm}^2/\text{min}$  in the direction of the thickness of said diffusion electrode.

10. The method for producing a membrane-electrode structure according to claim 1, wherein said polymer electrolyte membrane is formed from a sulfonated polyarylene based polymer solution, and when said catalyst paste comprises catalyst particles

consisting of a catalyst supported by carbon particles, an organic solvent solution containing a perfluoroalkylene sulfonic acid polymer, and a pore-forming material,

    said electrode catalyst layer is dried to such an extent that the content of the solvent becomes 20% or less by weight based on the total weight of said electrode catalyst layer, and the dried electrode catalyst layer is then thermally transferred onto and connected to said polymer electrolyte membrane.

11. The method for producing a membrane-electrode structure according to claim 10, wherein said thermal transfer is carried out under a pressure within the range of 1 to 5 MPa.

12. The method for producing a membrane-electrode structure according to claim 10, wherein said catalyst paste is applied on said sheet substrate whose surface has a contact angle to water of 55° to 105°.

13. The method for producing a membrane-electrode structure according to claim 10, wherein when said diffusion electrode is laminated on each electrode catalyst layer and they are then pressed under heating, the applied pressure is set within the range of 0.5 to 4 MPa.

14. A polymer electrolyte fuel cell having a membrane-electrode structure obtained by a production method comprising the steps of:

applying on a sheet substrate a catalyst paste comprising of an electron conducting material supporting a catalyst and an ion conducting material, and drying the same, so as to form an electrode catalyst layer;

thermally transferring said electrode catalyst layer onto each side of a polymer electrolyte membrane, so as to form a laminated body such that said electrode catalyst layer is connected to each side of said polymer electrolyte membrane;

applying on a carbon substrate layer a first slurry comprising a water-repellent material and an electron conducting material and drying the same to form a water-repellent layer, and then applying on said water-repellent layer a second slurry comprising an electron conducting material and an ion conducting material and drying the same to form a hydrophilic layer, so as to form a diffusion electrode consisting of said carbon substrate, said water-repellent layer and said hydrophilic layer; and

laminating the previously formed diffusion electrode on said electrode catalyst layer of said laminated body through said hydrophilic layer, pressing the two under heating, so as to integrate said laminated body and said diffusion electrode.

15. An electrical apparatus, which uses a polymer electrolyte fuel cell having a membrane-electrode structure obtained by a production method comprising the steps of:

applying on a sheet substrate a catalyst paste comprising of an electron conducting material supporting a catalyst and

an ion conducting material, and drying the same, so as to form an electrode catalyst layer;

thermally transferring said electrode catalyst layer onto each side of a polymer electrolyte membrane, so as to form a laminated body such that said electrode catalyst layer is connected to each side of said polymer electrolyte membrane;

applying on a carbon substrate layer a first slurry comprising a water-repellent material and an electron conducting material and drying the same to form a water-repellent layer, and then applying on said water-repellent layer a second slurry comprising an electron conducting material and an ion conducting material and drying the same to form a hydrophilic layer, so as to form a diffusion electrode consisting of said carbon substrate, said water-repellent layer and said hydrophilic layer; and

laminating the previously formed diffusion electrode on said electrode catalyst layer of said laminated body through said hydrophilic layer, pressing the two under heating, so as to integrate said laminated body and said diffusion electrode.

16. A transport machine, which uses a polymer electrolyte fuel cell having a membrane-electrode structure obtained by a production method comprising the steps of:

applying on a sheet substrate a catalyst paste comprising an electron conducting material supporting a catalyst and an ion conducting material, and drying the same, so as to form an electrode catalyst layer;

thermally transferring said electrode catalyst layer onto each side of a polymer electrolyte membrane, so as to form a laminated body such that said electrode catalyst layer is connected to each side of said polymer electrolyte membrane;

applying on a carbon substrate layer a first slurry comprising a water-repellent material and an electron conducting material, and drying the same to form a water-repellent layer, and then applying on said water-repellent layer a second slurry comprising an electron conducting material and an ion conducting material, and drying the same to form a hydrophilic layer, so as to form a diffusion electrode consisting of said carbon substrate, said water-repellent layer and said hydrophilic layer; and

laminating the previously formed diffusion electrode on said electrode catalyst layer of said laminated body through said hydrophilic layer, pressing the two under heating, so as to integrate said laminated body and said diffusion electrode.